NO_x-emissions from flameless coal combustion in air, Ar/O_2 and CO_2/O_2

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Abstract

Flameless or 'mild' combustion is a well known measure to reduce NO_x-emissions. This work aims at the application of the technique to coal combustors. Experiments have been carried out with lignite as well as bituminous coals showing an overall NO_x-reduction capability with the current burner design of about 20–50% depending on fuel type and the stoichiometry at the burner. The most important design feature is a high momentum of the combustion air inducing a strong recirculation which reduces the temperature variations and temperature peaks in the combustion chamber and thus prevents thermal NO formation. An investigation of NO_x-emissions under Ar/O_2 as well as CO_2/O_2 atmospheres was carried out in order to quantify the ratio of fuel- to thermal-NO. This investigation showed a high reduction of thermal NO in the flameless combustion mode. It also showed an increase of fuel-NO which was primarily related to the decrease of the peak flame temperature in flameless combustion. The intensive mixing in flameless combustion also allows a reduction of the oxidiser oxygen concentration under oxyfuel conditions to 15 vol % as compared to 17 vol % with a standard swirl burner.
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1. Introduction

In recent years much effort has been put into the reduction of NO_x-emissions from combustion processes. Secondary measures, for example DeNOx facilities, had to be introduced in order to comply with environmental restrictions. Primary measures to prevent NO_x-formation have been developed in the past and are still subject of investigation.

Among the best known strategies is air staging with which a reducing atmosphere is formed locally in the burner proximity that prevents NO_x-formation due to the lack of oxygen.

Another option consists of an additional burner level in which fuel is injected into the furnace after most of the combustion took place. In this case NO_x which has formed in the primary reaction zone can be reduced again by reaction with carbonaceous radicals in the reburning zone.

A concept based on high flue gas recirculation rates has been successfully proven to significantly reduce NO_x-emissions in gas and oil burners. The

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recirculating flue gas smoothes the temperature field within the furnace by mixing with the reactants. Due to this dilution the adiabatic flame temperature of the reacting mixture decreases and thus peak temperatures within the furnace are reduced. In literature this type of combustion is often called 'flameless oxidation' since no visible flame front develops [1]. Further commonly used names are 'mild combustion' [2,3] or if high air preheating is used 'high temperature air combustion' (HTAC, HiTAC) [4,5].

In the early 1990s first experimental investigations on high temperature air combustion of coal were carried out at the IFRF where the aim was to explore synergies from the combined use of gas turbines and coal combustion. Therefore, a gas turbine exhaust gas with reduced oxygen content was used for the combustion of coal. These experiments were conducted with conventional burners [6].

Towards the end of the 1990s the IFRF carried out combustion tests at a thermal load of 580 kW with a completely new injection scheme, separating the coal jet from the highly preheated (1350 °C) combustion air [7]. A significant reduction in NO_x-emissions could be achieved. Just recently this burner concept has been revitalised and based on the previous results a complete furnace has been designed numerically [8]. This design aims at high recirculation rates within the whole furnace, thus encouraging reburning.

In 2001 an investigation of flameless combustion was carried out at RWTH Aachen University (RWTH) showing the applicability to pulverised coal combustion under pressurised conditions [9]. The thermal load based on coal flow was 100 kW without additional combustion air preheating. The experiments were carried out at a furnace pressure of 3 bar (abs). This integrated burner design also proved to significantly reduce NO_x-emissions in coal combustion.

From 2005 on an integrated burner design has been developed within the European Union supported project FLOX-COAL. The aim is to obtain a burner which is suitable for retrofitting old power plants as well as for the design of new power plants. The NO_x-emissions with these burners shall be low enough so that DeNOx facilities will become obsolete.

This paper describes the combustion behaviour of this new burner design under air staging conditions. Especially the NO_x-emissions in dependence of the stoichiometric ratio at the burner has been determined for different coals. In order to identify the mechanisms for NO_x-production and reduction, experiments have also been conducted in argon/oxygen and in carbon dioxide/oxygen atmospheres. Thereby preventing thermal NO, the fraction of total NO that originates from fuel bound nitrogen has been determined.

2. Experimental setup

The burner design was tested in two geometric similar test rigs at RWTH and at the University of Stuttgart (USTUTT). The test rigs are vertical, cylindrical, top fired furnaces which employ electrical heating for regulation of the wall temperature.

Pilot scale experiments were performed at the pulverised coal combustion facility at RWTH. A sketch of the facility is shown in Fig. 1. The diameter of the furnace is 400 mm and the total length 4200 mm. In comparison the lab-scale facility at USTUTT has a diameter of 200 mm at a total length of 2500 mm.

A burner was developed which is closely related to the design of flameless gas burners. The predominant design criterion for the pulverised coal burner is the avoidance of high temperature zones in order to reduce the formation of thermal NO after the Zeldovich mechanism.

According to [10] a high potential for NO-reduction is also given by deferring the mixing between coal and combustion air. Thereby NO formation after the volatile-NO path, which makes up a large portion of the total fuel-NO, can be reduced.
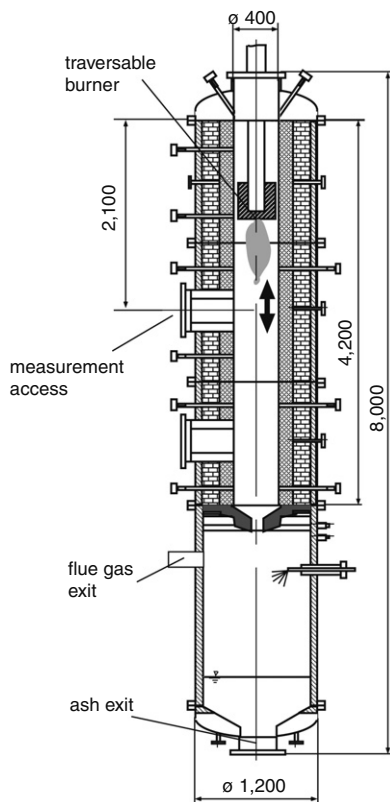

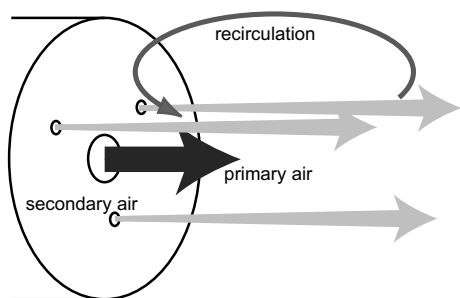Fig. 1. Test facility at RWTH Aachen University.

Fig. 2. Sketch of burner design and flow scheme.

In accordance with gas burners this is achieved by good mixing of the unburnt mixture and the flue gases. A high inlet momentum of the combustion air is used to induce the required recirculation of the flue gases which are then entrained into the fresh gases and the coal. A sketch of the investigated burner design is shown in Fig. 2. The coal is transported by a carrier air flow and enters the chamber through a central inlet with a velocity of about 10 m/s. Exceptions are experiments labelled L1 where the carrier air flow was 14 m/s and experiments labelled L2 where pure nitrogen was used as coal carrier. For the data labelling and experimental boundary conditions see Table 1. The combustion air is injected through three nozzles which are annularly arranged around the central coal jet. At RWTH the stoichiometric

ratio at the burner was varied by adjusting the air flow at constant coal flow whereas at USTUTT the opposite procedure was used. Thus the thermal load in pilot scale experiments was constant at 40 kW and in lab-scale experiments varied between 5 and 8 kW.

Proximate and ultimate analysis of the coals referred to in Table 1 are given in Table 2.

Additional burnout air was introduced downstream of the main reaction zone. In case of the pilot scale facility at RWTH this burnout air was introduced at an axial distance of 2000 mm and in case of the lab-scale facility at USTUTT at an distance of 1800 mm from the burner level.

Due to the arrangement of the nozzles recirculated hot flue gases not only dilute the combustion air but can also penetrate between the air jets directly to the coal jet where the particles are thereby heated in a reducing atmosphere (indicated in Fig. 2). An additional advantage of the recirculation is that NO formed during combustion is to some extent brought into contact with volatile radicals in the absence of oxygen. This means that with flameless combustion NO-reduction occurs not only due to the reduction of thermal NO due to reduced temperature peaks but that there is also another benefit that is similar to reburning systems. According to [11] conventional reburning systems can have a NO_x-reduction efficiency between 50% and 70% so that there is a high potential for flameless combustion.

Table 1
Flow parameters

Key	Furnace	Coal	Coal flow (kg/h)	Wall temperature	Secondary air velocity	Preheating
L1[*]	RWTH	LL	6.4–6.7	900 °C	100–200 m/s	None
L2[**]	RWTH	LL	6.4–6.7	900 °C	100–200 m/s	None
L3	RWTH	LL	6.4–6.7	900 °C	100–200 m/s	None
L4	USTUTT	LL	0.93–1.32	1300 °C	100 m/s	300 °C
L5	USTUTT	LL	1.26–1.44	1100 °C	140 m/s	300 °C
L6	USTUTT	LL	1.02–1.17	1100 °C	100 m/s	300 °C
L7	USTUTT	LL	0.98–1.15	1000 °C	100 m/s	300 °C
L8	USTUTT	LL	1.23–1.43	1000 °C	140 m/s	300 °C
L9	USTUTT	LL	0.9–1.3	1100 °C	120 m/s	300 °C
LR	USTUTT	LL	0.9–1.3	1300 °C	Flame burner	300 °C
B1	RWTH	PB	4.2	850 °C	40–130 m/s	None
B2	USTUTT	KK	0.82–1.17	1300 °C	100 m/s	300 °C
B3	USTUTT	KK	0.95–1.16	1100 °C	100 m/s	300 °C
B4	USTUTT	KK	1.01–1.16	1300 °C	100 m/s	300 °C
B5	USTUTT	BU	0.97–1.07	1300 °C	100 m/s	300 °C
B6	USTUTT	BU	0.91–0.96	1100 °C	120 m/s	300 °C
BR	USTUTT	PB	0.61–0.87	1300 °C	Flame burner	300 °C
LA1	USTUTT	LL	1.03	1300 °C	100 m/s	300 °C
LA2	USTUTT	LL	1.03	1300 °C	Flame burner	300 °C
LA3	USTUTT	LL	1.22	Var.	100 m/s	300 °C
LO1	RWTH	RL	5.6	900°	200–280 m/s	None
LO2	RWTH	RL	8.4	850°	100–130 m/s	None
LO3	RWTH	RL	5.6	850°	Flame burner	None

[*], carrier air velocity 14 m/s; [**], coal carrier nitrogen; L, lignite; B, bituminous; R, reference; A, experiments in Ar/O_2 atmosphere; O, experiments in CO_2/O_2 atmosphere.

Table 2
Proximate and ultimate analysis in [mass %] of coals used during experiments (LL, Lusatian lignite; RL, Rhenish lignite; PB, bituminous coal blend from Poland; KK, bituminous coal Kleinkopje; BU, bituminous coal Budryk)

	LL	RL	PB	KK	BU
Water	10.20	8.40	0.60	3.60	6.90
Ash	4.90	4.10	3.60	18.60	26.30
Vol.	48.70	46.60	33.00	21.60	23.30
Char	36.20	40.90	62.80	56.20	43.20
C	56.70	67.40	85.90	65.30	57.30
H	5.60	4.24	5.09	3.90	4.00
O	21.44	14.70	2.94	7.02	3.52
N	0.55	0.86	1.41	1.30	0.77
S	0.61	0.30	0.46	0.28	0.64

3. Results and discussion

3.1. Results from combustion in air

In Fig. 3 the NO_x-emissions for Lusatian lignite measured in the flue gases and calculated at 6%-oxygen are given versus the stoichiometric ratio at the burner. Data labelling is in accordance to the key in Table 1. Besides the general well known trend to higher NO_x-emissions for higher burner excess air ratios a broad variation of the data from the two test facilities can be seen. However, in spite of the different schemes used for the variation of the stoichiometric ratio, results from both test facilities show similar characteristics. Therefore, the combustion scheme can be considered stable to variations in the boundary conditions.

For a comparison to 'normal' or 'flame' combustion a conventional jet burner has been used

at USTUTT in order to collect reference data. The boundary conditions in the reference experiments, labelled 'LR', were equal to those in flameless combustion labelled 'L4'. Between a stoichiometric ratio at the burner of 0.8 and 0.9, data from flameless combustion shows lower NO_x-emissions. For higher stoichiometric ratios flameless combustion yields increased NO_x-emissions.

For a particular setting of the boundary conditions (wall temperature and flow rates) it is possible to stay below the regulatory limit of 200 mg/m^3 with flameless combustion despite an increase of the stoichiometric ratio at the burner. The best values that could be obtained are 0.9 in the pilot scale facility and 0.95 in the lab-scale facility.

The corresponding graph for bituminous coals is given in Fig. 4. The general tendency is the same as for lignite, the actual values are slightly higher. In the pilot scale facility it was possible to increase the stoichiometric ratio at the burner for bituminous coal to 0.7 and in the lab-scale facility to 0.9 while staying below the regulatory limit.

From the experience with gas burners it can be assumed that the reduction of NO_x-emissions is due to the avoidance of high temperature regions. The good mixing induced by the high momentum is responsible for a homogenisation of the temperature field within the furnace.

Velocity measurements conducted at RWTH show the extensive recirculation zone next to the strong central jet. An exemplary plot at an axial distance of 100 mm from the burner plane is shown in Fig. 5. The measurement plane was slightly inclined to the plane between the secondary air inlet and the coal inlet. Thus the peak velocity of about 32 m/s measured at a radial distance of 10 mm indicates the boundary of a
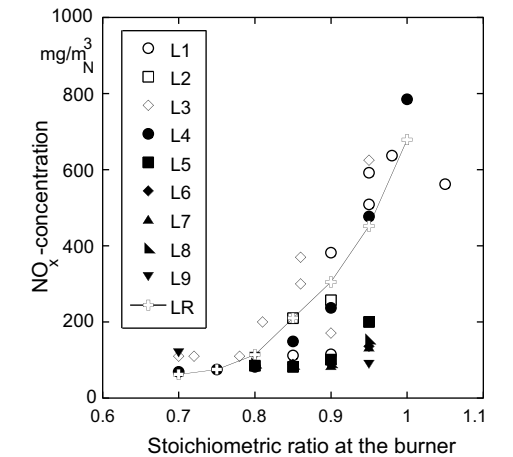

Fig. 3. NO_x-emissions over burner excess air ratio for lignite, data key in Table 1, open symbols, RWTH; filled symbols, USTUTT.
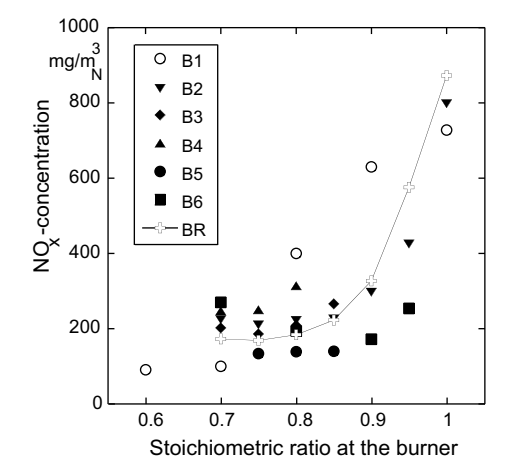

Fig. 4. NO_x-emissions over burner excess air ratio for bituminous coal, data key in Table 1, open symbols, RWTH; filled symbols, USTUTT.
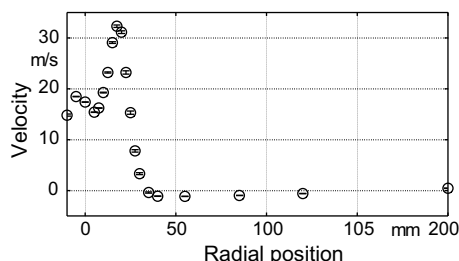
Fig. 5. Profile of axial velocity at a distance of 100 mm from the burner plane, data key 'B1' in Table 1, stoichiometric ratio at the burner: 0.7, secondary air velocity: 70 m/s.

secondary air jet. The central coal jet on the axis has been accelerated from about 10 m/s at the inlet to around 19 m/s at 100 mm distance. Between the two jets a local minimum can be distinguished showing that the two jets have not merged yet. From 35 mm outwards a recirculation of flue gases extends almost to the furnace walls with a velocity of about 1 m/s. The measurement at 200 mm radial position is influenced by wall effects. Confidence intervals for the measurements are given but are, due to the large number of evaluated particles, negligible.

3.2. Results from combustion in Ar/O_2-atmosphere

In order to identify the origin of NO-reduction, experiments have been carried out at USTUTT using Lusatian lignite in an argon/oxygen atmosphere. The oxygen content by volume was held constant to the one in air (21 vol %). Due to the absence of molecular nitrogen thermal NO as well as prompt NO cannot be formed in this atmosphere. Thus the experiments reflect fuel-NO-concentration. Figure 6 shows the NO-concentration in argon/oxygen atmosphere along
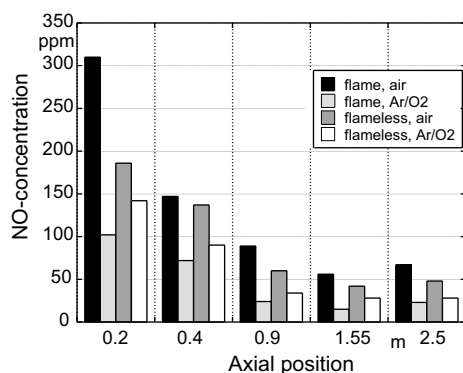

Fig. 6. NO-concentration along the centreline of the furnace in Ar/O_2 atmosphere and air for flame and flameless combustion mode under air staging conditions, stoichiometric ratio at the burner 0.8, data keys 'LA1' and 'LA2' in Table 1.

the centreline of the furnace for a jet flame and flameless combustion at a stoichiometric ratio at the burner of 0.8. For each measurement a reference value from air combustion is given.

All experiments show a decrease of the NO-concentration from the burner vicinity to the exit. Between 1550 and 2500 mm a minor increase can be identified due to the burnout air (Ar/O_2, respectively) which is introduced at 1800 mm.

If the experiments in air are considered, NO-emissions from flameless combustion are lower than those in flame combustion. This was already discussed (Fig. 3). In the burner vicinity the NO-concentration for flameless combustion is about half of the concentration for flame combustion and in the exit it is still about 25% lower.

Experiments in argon/oxygen atmosphere show the converse trend. Fuel-NO-concentrations from flameless combustion exceed the values obtained from flame combustion.

The difference in NO-concentration between air and argon/oxygen atmosphere can be considered thermal NO. In flameless combustion this thermal NO-concentration is about half the value from flame combustion for all data throughout the furnace. From that it can be concluded that thermal NO is significantly curbed with flameless combustion.

In flameless combustion of Lusatian lignite fuel-NO is responsible for about 60% of total NO whereas in flame combustion it is 35%.

On the other hand, the difference between flame and flameless combustion in argon/oxygen atmosphere indicates that fuel-NO is slightly increased with flameless combustion. This increase may be due to a reduced maximum temperature in flameless combustion. The negative dependency of fuel-NO on combustion temperature was already indicated by [12,13] and confirmed for the current setup in flameless combustion experiments at different wall temperatures, Fig. 7. For all axial distances data taken at the lower wall temperature shows a higher NO-concentration. A side-effect of this temperature dependence is that the actual fuel-NO yields can be underpredicted due to the higher adiabatic flame temperature in the argon/oxygen atmosphere as compared to air.

To to further investigate the influence of the flame temperature, experiments have also been conducted in an atmosphere of carbon dioxide and oxygen. These oxyfuel experiments have been carried out RWTH.

3.3. Results from combustion in CO_2/O_2-atmosphere

The oxygen concentration and hence the adiabatic flame temperature was varied. In Fig. 8 data from flameless and flame combustion are given. The flame burner, a standard swirl burner
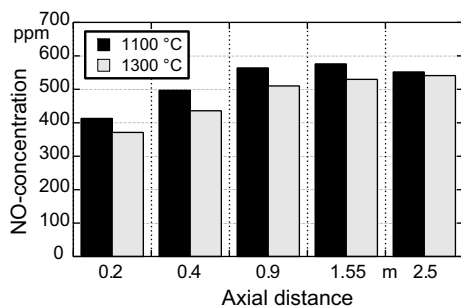
Fig. 7. Fuel-NO-concentration along the centreline measured at different wall temperatures, unstaged flameless combustion, data key 'LA3' in Table 1.

described in [14], was operated between 17 and 26 vol % oxygen in the oxidiser. Below 17 vol % no stabilised combustion was possible with this burner. However, due to the good mixing within the furnace flameless combustion was stable down to 15 vol % oxygen in the oxidiser. At this concentration the CO-concentration in the flue gases was 39 mg/m$_N^3$ still indicating good burnout for the flameless burner 'A' described above. The second flameless burner, a single nozzle burner which was at first designed for the operation under pressurised conditions (described in [9]), produced higher CO-emissions at all oxygen concentrations.

In order to eliminate the influence of the dilution and the increased flue gas volume flow at lower oxidiser oxygen concentrations, NO_x-emissions are not given as flue gas concentrations but are calculated in terms of total fuel-nitrogen conversion.

Experiments with the swirl burner show the same behaviour as the argon/oxygen experiments.
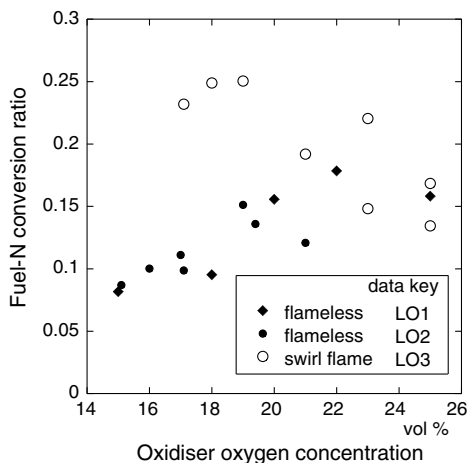

Fig. 8. Fuel-N conversion ratio versus oxidiser oxygen concentration in CO_2/O_2 atmosphere for flame and flameless combustion, data key in Table 1.

The NO_x-emissions increase as the flame temperature decreases due to the dilution of the oxidiser with CO_2. This finding is contrary to the conclusion found by [15]. However, the experiments in [15] were conducted at higher oxygen concentrations, i.e. 21 and 30 vol %, and with a coal with almost twice the nitrogen content (1.62 mass % compared to 0.86 mass %). At 21 vol % a fuel-N conversion of around 30% is reported which is in the same order of magnitude as the current 20% at 21 vol % oxygen. The increased conversion of almost 80% reported at 30 vol % oxygen in oxidiser indicates a different effect. At high oxygen concentrations the abundance of oxygen yields higher NO conversion whereas at the low oxygen concentrations of the current investigation the decreased flame temperature is made responsible for the increased NO production.

Experiments conducted with the flameless burners show the opposite trend. With both burners NO_x-emissions slightly decrease towards lower oxygen concentrations and thus lower flame temperature. The absolute values of both burners are comparable despite the different thermal load.

The reduced NO_x-emissions can be attributed to improved mixing due to a higher secondary air velocity at lower oxidiser oxygen concentrations. The recirculation is apparently intensified causing NO_x to be brought back to the primary reaction zone. Due to the mixing the local oxygen concentration drops well below the inlet concentration so that NO_x-formation is to some extent prevented and reburning of recirculated NO_x is improved.

However, the velocity in the oxyfuel experiments is too high for practical combustors since the burner geometry has not been adapted to the bigger volume flow rate at low oxygen concentrations.

It can be concluded that the conversion of fuel-NO is strongly temperature dependent and that it is possible to compensate for that with flameless combustion. The reburning effect of flameless combustion can be improved by a higher momentum of the secondary air jets as in the case of reduced oxygen concentrations.

3.4. Burnout and CO-emission of flameless combustion

Table 3 shows measurements of the CO-concentration in the flue gases as well as results from ash analyses. For most experiments at the bench scale facility at USTUTT ash analyses were carried out. In ash samples from lignite combustion no measurable quantities of carbon, hydrogen or nitrogen were detected. From these results complete burnout with flameless combustion under air-staged conditions can be concluded. Carbon in ash for the bituminous coal experiments shows a dependence on the wall temperature. With

Table 3
Burnout measurements in [mass%] and CO-concentration in [mg/m$_N^3$] at 6% oxygen in the flue gases for different settings

	L1	L2	L6	L7	L8	L9	B2	B3	B5	B5*	B5
SR	0.90	0.90	0.85	0.85	0.85	0.85	0.75	0.75	0.75	0.8	0.85
Carbon	—	—	bdl	bdl	bdl	bdl	2.3	18.9	—	0.76	—
Hydrogen	—	—	bdl	bdl	bdl	bdl	bdl	bdl	—	bdl	—
Nitrogen	—	—	bdl	bdl	bdl	bdl	bdl	0.38	—	bdl	—
Sulphur	—	—	2.7	2.8	2.1	2.5	0.37	1.10	—	0.8	—
CO	6	2	29	36	29	29	44	78	33	35	34

SR, stoichiometric ratio at the burner; bdl, below detection limit (<0.3); —, not measured; *, burnout air during ash sampling at 1000 mm from burner.

increasing wall temperature carbon conversion is significantly enhanced. At the lower temperature (1100 °C) carbon in ash is as high as 18.9 mass % whereas at 1300 °C carbon in ash is only 2.3 mass %. However, further experiments without air staging showed that the burnout is enhanced and less carbon remains in the ash.

Throughout all the experiments conducted during the current investigation CO-emissions were well below the regulatory limit of 100 mg/m^3 and in the case of lignite even below 50 mg/m$_N^3$ with a minimum of 2 mg/m$_N^3$ measured in the experiments at RWTH. The comparison of experiments carried out at RWTH and those at USTUTT indicates that burnout as defined by the CO-concentration at the larger furnace is improved. This is due to the longer residence time after the addition of burnout air.

4. Conclusion

Due to several advantages in terms of NO$_x$-emissions flameless combustion is a promising technology for retrofitting old or designing new furnaces.

- flameless combustion is achieved by a strong internal recirculation which causes the combusting mixture to be diluted and thus thermal NO to be avoided
- minimum staging in order to stay below regulatory limits of 200 mg/m$_N^3$ can be reduced to about 0.9 for lignite and 0.8 for bituminous coal
- CO-emissions in flameless combustion of lignite are as low as 2–3 mg/m$_N^3$, for bituminous coal still below 50 mg/m$_N^3$.

Experiments in nitrogen-free atmospheres revealed fuel-NO behaviour in flameless combustion:

- experiments of flameless combustion in argon/oxygen atmosphere revealed thermal NO$_x$-emissions to be cut by about one half compared to flame combustion

- these experiments also show a slight increase in fuel-NO-concentration for flameless combustion as compared to flame combustion which can be attributed to a lower combustion temperature
- experiments with a conventional swirl burner in CO$_2$/O$_2$ atmosphere show an increase in NO$_x$-emissions with a decrease in oxidiser oxygen concentration
- flameless combustion under oxyfuel conditions shows a slight decrease in NO$_x$-emission with a decrease in oxidiser oxygen concentration
- the decrease in NO$_x$-emission at lower oxidiser oxygen concentrations and hence lower peak temperatures can be due to the intensified recirculation which brings flue gases back to the primary reaction zone where NO$_x$ can be effectively reburned
- comparison of the trends with the swirl burner and flameless burners gives rise to the conclusion, that the reductive effect of mixing on NO$_x$-emissions is more dominant than the increasing effect of reduced maximum combustion temperature
- with flameless combustion under oxyfuel conditions it is possible to reduce the oxidiser oxygen content to as little as 15 vol %.

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